COBEM 2021
26th International Congress of Mechanical Engineering

ABCM
Associação Brasileira de Engenharia e Ciências Mecânicas
November 22-26, 2021, Virtual Congress, Brazil

COB-2021-1428

CONTINUOUS MODEL FOR DISSOLUTION IN ELASTIC SOLIDS UNDER TENSION

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Abstract. *In this paper, we present an analytical description of the effective dissolution kinetics of spherical particles into a solvent. We investigate the role of stress on the dissolution of elastic solids in liquids. Towards this end, we first formulate a continuum theory to describe the solid-solution system made up of an elastic solid separated from the surrounding solution by a sharp boundary that evolves due to dissolution. In addition to the usual balances needed to describe this kind of system, we introduce an extra force balance, the so-called configurational force balance, from which we derive a general saturation condition at the solid-solution interface. Under suitable constitutive assumptions, we arrive at two sets of equations to be solved in moving domains: the equations of linear elasticity on the dissolving solid and the diffusion equation to be solved for the solid content in the solution. These equations are coupled since their domains are separated by a moving boundary whose evolution is governed by a stress-dependent dissolution rate and saturation condition. The latter provides the boundary condition for the diffusion equation in the moving boundary. Finally, consider analytical and numerical examples to illustrate the use of the proposed model.*

Keywords: Dissolution, Configurational Forces, Corrosion, Drug Delivery

1. INTRODUCTION

Chemical, mechanical, electric, and environmental phenomena are some of the phenomena that can influence on corrosion process. It is a challenge to understand how each one influences the process because, in general, they act together. Then, the interest in corrosion modeling is increasing to well understand the corrosion process and contribute to the reduction of failures. The fundamental mechanism behind the corrosion process is the dissolution (Sarkar *et al.*, 2012). The work of (Nguyen *et al.*, 2017) and (Mai and Soghrati, 2017) relate the pit corrosion with the mechanical problem using phase field model. Phase field model presented at (Mai *et al.*, 2016) was used by (Mai and Soghrati, 2018) that incorporated electrical potential effects.

In this paper, we rederive Gibbs's relation that governs the local thermodynamic equilibrium of a stressed pure solid in contact with a solution phase. We propose that using configurational forces balance. Then, the manner in which it has been derived here differs from the classical variational approach of Gibbs and from the approach proposed by (Lehner and Bataille, 1984).

2. THEORY

2.1 Problem Definition

We consider a material body (R_t) composed of a solid phase (R_t^+) and a liquid phase (R_t^-). These phases are separated by an interface (S_t), metal ions migrate from R_t^+ to R_t^-. To formulate this problem, we need select a region D_t like Figure 1. To simplify the problem, we assume some characteristics like consider an isothermal problem where the diffusion of ions in the metallic phase is negligible.

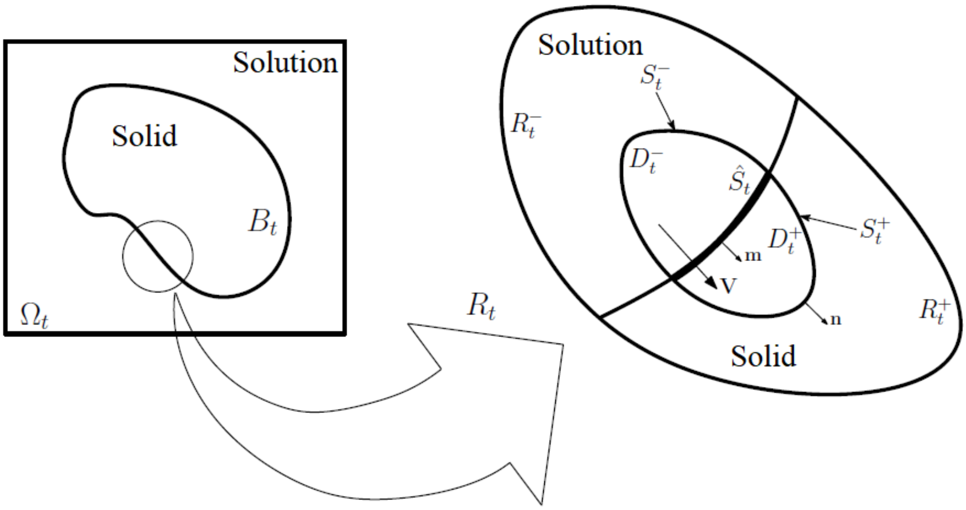


Figure 1. An arbitrary subregion D_t that intersects the interface S_t. $D_t = D_t^+ \cup D_t^-$. The segment S_t contained in D_t is called $\hat{S}_t$. Note that $\partial D_t = S_t^+ \cup S_t^-$, $\partial D_t^+ = S_t^+ \cup \hat{S}_t$ and $\partial D_t^- = S_t^- \cup \hat{S}_t$. The unit vector $\mathbf{n}$ is normal to ∂D_t and $\mathbf{m}$ is normal to interface S_t. And V is the speed with which the interface evolves. Figure adapted from (Abeyartne, 2012)

2.2 Balance Equations

Three regions are considered on the problem: solid, solution and interface. We propose balance equations for each region and localize them by applying the transport and divergence theorem.

2.2.1 Solid

The solid is comprehended by the region D_t^+ of the Figure 1. Localized equations are described by:

$$\dot{\rho_S}(\mathbf{x},t) + \rho_S div\mathbf{v}(\mathbf{x},t) = 0 \tag{1}$$

$$div\mathbf{T}_S(\mathbf{x},t) + \mathbf{b}(\mathbf{x},t) = 0 \tag{2}$$

$$\mathbf{T}_S(\mathbf{x},t) = \mathbf{T}_S^T(\mathbf{x},t) \tag{3}$$

$$\dot{\overline{\rho_S(\mathbf{x},t)c_S(\mathbf{x},t)}} + \rho_S(\mathbf{x},t)c_S(\mathbf{x},t)div\mathbf{v}(\mathbf{x},t) = 0 \tag{4}$$

$$\rho_S(\mathbf{x},t)\dot{\psi_S}(\mathbf{x},t) - \mathbf{T}^+(\mathbf{x},t)\cdot\mathbf{D}(\mathbf{x},t) \leq 0 \tag{5}$$

where ρ_S is the density of the solid, $\mathbf{x}$ is the position, t is the time, $\mathbf{T}^+$ is the stress tensor, $\mathbf{b}$ is the generalized body force that is described by $\mathbf{b} = \mathbf{b_o} - \rho_S\dot{\mathbf{v}}$. And b_o is the body force that act on the body, $\mathbf{n}$ is the unit vector normal to ∂D_t, c_S is the mass fraction, ψ_S is the Helmholtz free energy and $\mathbf{v}$ is the barycentric velocity.

2.2.2 Solution

The solution is comprehended by D_t^- of the Figure 1 and is composed of two elements: the liquid and dissolved ions. The localized equations are described by:

$$\dot{\rho_F}(\mathbf{x},t) + \rho_F div\mathbf{v}(\mathbf{x},t) = 0 \tag{6}$$

$$div\mathbf{T}_F(\mathbf{x},t) + \mathbf{b}(\mathbf{x},t) = 0 \tag{7}$$

$$\mathbf{T}_F(\mathbf{x},t) = \mathbf{T}_F^T(\mathbf{x},t) \tag{8}$$

$$\dot{\overline{\rho_F(\mathbf{x},t)c_{SL}(\mathbf{x},t)}} + \rho_F(\mathbf{x},t)c_{SL}(\mathbf{x},t)div\mathbf{v} + div\mathbf{J}_{SL}(\mathbf{x},t) = 0 \tag{9}$$

$$\dot{\overline{\rho_F(\mathbf{x},t)c_L(\mathbf{x},t)}} + \rho_F(\mathbf{x},t)c_L(\mathbf{x},t)div\mathbf{v}(\mathbf{x},t) + div\mathbf{J}_L(\mathbf{x},t) = 0 \tag{10}$$

$$\begin{aligned}\dot{\overline{\rho_F(\mathbf{x},t)\psi(\mathbf{x},t)}} + \rho_F(\mathbf{x},t)\psi(\mathbf{x},t)div\mathbf{v}(\mathbf{x},t) + \mu_{SL}(\mathbf{x},t)div\mathbf{J}_{SL}(\mathbf{x},t) + \mathbf{J}_{SL}(\mathbf{x},t)grad\mu_{SL}(\mathbf{x},t) +\\ \mu_L(\mathbf{x},t)div\mathbf{J}_L(\mathbf{x},t) + \mathbf{J}_L(\mathbf{x},t)grad\mu_L(\mathbf{x},t) - \mathbf{v}(\mathbf{x},t)div\mathbf{T}(\mathbf{x},t) - \mathbf{b}_o(\mathbf{x},t)\cdot\mathbf{v}(\mathbf{x},t) +\\ \rho_F(\mathbf{x},t)\dot{\mathbf{v}}(\mathbf{x},t)\cdot\mathbf{v}(\mathbf{x},t) - \mathbf{T}^-(\mathbf{x},t)\cdot grad\mathbf{v}(\mathbf{x},t) \leq 0\end{aligned} \tag{11}$$

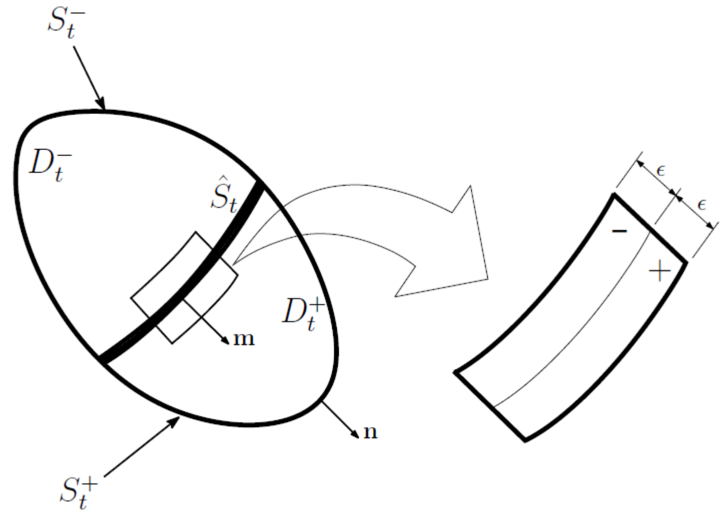


Figure 2. Representation of a *pilbox* with thickness ϵ on interface

where ρ_F is the density of the solution, $\mathbf{T}_F$ is the stress tensor, c_{SL} is the mass fraction of dissolved metal ions, $\mathbf{J}_{SL}$ is the ions flux, c_L is the mass fraction of liquid, $\mathbf{J}_L$ the liquid flux, μ_{SL} is the chemical potential of dissolved metal ions and μ_L the liquid chemical potential.

2.2.3 Interface

The interface is represented by S_t (Figure 1). Metallic ions migrate from solid to the solution, then the interface change with time. So, we can consider a new degree of freedom. To describe this new degree of freedom we use configurational forces. As such, in addition to the balance equation of mass, linear and angular momentum, transport, and second law we include the balance of configurational forces (Gurtin, 1999).

$$f^i(\mathbf{x},t) + f^e(\mathbf{x},t) = 0, \tag{12}$$

Where f^i is an internal configurational force and f^e is an external configurational force.

Looking Figure 2, we have $D_t = D_t^+ \cup D_t^-$, $\partial D_t = S_t^+ \cup S_t^-$, $\mathbf{m}$ is the normal vector to $\hat{S}_t$. The equations on interface, considering a thickness ϵ are:

$$[\rho(V - \mathbf{v}\cdot\mathbf{m})] = 0 \tag{13}$$

$$[\rho\mathbf{v}(V - \mathbf{v}\cdot\mathbf{m}) + \mathbf{Tm}] = 0 \tag{14}$$

$$[\rho c(V - \mathbf{v}\cdot\mathbf{m})] = -\mathbf{J_F}\cdot\mathbf{m} \tag{15}$$

$$-[\rho\psi(V - \mathbf{v}\cdot\mathbf{m})] - [\mathbf{Tm}\cdot\mathbf{v}] - (\mu_{SL}\mathbf{J}_{SL}\cdot\mathbf{m} + \mu_L\mathbf{J}_L\cdot\mathbf{m}) + f^i\cdot(V - \mathbf{v}\cdot\mathbf{m}) \leq 0 \tag{16}$$

Where $\mathbf{V}$ is the velocity of the interface (represented on Figure 1)

2.3 Constitutive Theory

To solve the problem, we develop a constitutive theory by making choices for each part of the body that allows us to find a solution to our variables.

2.3.1 Solid

We assume a linear elastic solid, a deformable solid and work with small depormations from which we get our constitutive equation:

$$\hat{\mathbf{T}}_S(\mathbf{F}) = \rho_S\mathbf{F}^T\frac{\partial\hat{\psi}(\mathbf{F})}{\partial\mathbf{F}} \tag{17}$$

$$\mathbf{F} = grad(\mathbf{x}).$$

2.3.2 Solution

We assume a compressible fluid, inviscid, binary, and diluted solution. We also assume a hydrostatic pressure p for the solution, then:

$$\mathbf{T}_F = -p\mathbf{I} \tag{18}$$

This assumption and the equation 11 allows the following conclusion:

$$\psi_F = \mu_{SL}c_{SL} + \mu_L c_L - \frac{p}{\rho_F}, \tag{19}$$

The equation 19 is an important Gibb's relation, where $\psi_F + \frac{p}{\rho_F} = g^F$ is the specific Gibb's potential Lehner and Bataille (1984). At binary solutions we have the following relations for mass fraction and flux:

$$c_{SL}(\mathbf{x},t) + c_L(\mathbf{x},t) = 1 \qquad \rightarrow \qquad c(\mathbf{x},t) = c_{SL}(\mathbf{x},t) = 1 - c_L(\mathbf{x},t) \tag{20}$$

$$\mathbf{J}_{SL}(\mathbf{x},t) + \mathbf{J}_L(\mathbf{x},t) = 0 \qquad \rightarrow \qquad \mathbf{J}(\mathbf{x},t) = \mathbf{J}_{SL}(\mathbf{x},t) = -\mathbf{J}_L(\mathbf{x},t), \tag{21}$$

Then we rewrite the equation 11:

$$-\dot{\rho_F}\frac{p}{\rho_F} + \rho_F\dot{\psi_F} + \mathbf{J}\cdot grad\mu - \rho_F\dot{c}\mu \leq 0 \tag{22}$$

Where $\mu = \mu_{SL} - \mu_L$ is the chemical potential of solid dissolved in a liquid related to the chemical potential of the liquid. We have to introduce constitutive relations for ψ_F, μ and $\mathbf{J}$. Then, guided by inequality 22, we propose the following:

$$\psi_F = \hat{\psi}_F(\rho_F, c, grad\mu) \tag{23}$$

$$\mu = \hat{\mu}(\rho_F, c, grad\mu) \tag{24}$$

$$\mathbf{J} = \hat{\mathbf{J}}(\rho_F, c, grad\mu) \tag{25}$$

This allows us to conclude that the flux $\mathbf{J}$ can be written as Fick's law.

$$\mathbf{J} = -\rho_F D grad c, \tag{26}$$

where D is the diffusion coefficient.

2.3.3 Interface

We assume that the interface movement is quasi-static and that do not exist shear force between the solid and fluid. As already indicated the hidrostatic pressure on solution is p, then $\mathbf{T}_F \cdot \mathbf{m} = -p\mathbf{m}$. Therefore, in equilibrium condition:

$$\mathbf{T}_S \cdot \mathbf{m} = -p\mathbf{m} \tag{27}$$

In particular,

$$[\mathbf{Tm}\cdot\mathbf{v}] = \dot{m}p[\frac{1}{\rho}] \tag{28}$$

Where,

$$\dot{m} = \rho(V - v\cdot n)^+ = \rho(V - v\cdot n)^- \tag{29}$$

using relations 21 and 15, the inequality 16 can be rewrited by:

$$-\dot{m}([\psi] + p\left[\frac{1}{\rho}\right] - (\mu_{SL} - \mu_L)[c] - f^i) \leq 0 \tag{30}$$

A constitutive choice for f^i, after some assumptions is:

$$f^i = \beta\dot{m} - ([\psi] + p[\nu] - (\mu_{SL} - \mu_L)[c]) \tag{31}$$

Where $\dot{m}$ is related to interface movement. By assuming that the configurational external forces are zero, by the force equilibrium f^i is also zero. And if the interface moves without energy dissipation, we conclude that:

$$[\psi] + p[\nu] - (\mu_{SL} - \mu_L)[c] = 0 \tag{32}$$

That is an equation usually used to study dissolution problems and can be rewritten, after some manipulation, like:

$$\psi_S + \frac{p}{\rho_S} = \mu_{SL} \tag{33}$$

This result was discovered by Gibbs in 1906 Lehner and Bataille (1984) and here we arrive at the same result using configurational forces balance. Then, it is possible to write the govern equation to interface evolution like:

$$\dot{m} = \frac{1}{\beta}(\psi_S + \frac{p}{\rho_S} - \mu_{SL}) \tag{34}$$

Using the solution theory, we can write the chemical potential (μ_{SL}):

$$\mu_{SL} = \mu^*_{SL}(p,T) + \frac{RT}{M_{SL}} ln\gamma_{SL}x_{SL} \tag{35}$$

Where x_{SL} is the mole fraction, γ_{SL} is the activity coefficient, R is the universal gas constant, M_{SL} is the molar mass, μ^*_{SL} is the reference chemical potential which does not depend on the concentration and T is the temperature.

2.4 Govern Equations

We solve the governing equations to find the dissolution rate of a material. If the body is submitted to mechanical stresses we solve also the elastic equation. In this section, we write the governing equations for the solid, solution, and interface.

2.4.1 Solid

We disregard the body forces and restrict our attention to small-strain. Then we solve the equations 2 and 4, that can be rewritten by:

$$div\mathbf{T}_S = 0 \tag{36}$$

$$\dot{c}_S = 0 \tag{37}$$

2.4.2 Solution

With relations 20, 21 and the expression 26, disregarding the fluid movement, we rewrite 9 and 10:

$$\dot{c} - D\nabla^2 c = 0 \tag{38}$$

2.4.3 Interface

It is convenient to discuss such relaxation process in terms of the evolution of an equilibrium chemical potential μ^e_{SL} and an equilibrium mole fraction x^e_{SL}, then:

$$\mu^e_{SL} = \mu^*_{SL}(p,T) + \frac{RT}{M_{SL}} ln\gamma^e_{SL}x^e_{SL} \equiv \psi_S + \frac{p}{\rho_S} \tag{39}$$

Then, it is possible to rewrite the equation of interface movement (34).

$$\dot{m} = \frac{1}{\beta}(\mu^e_{SL} - \mu_{SL}) = \frac{RT}{\beta M_{SL}} \frac{\gamma^e_{SL}x^e_{SL}}{\gamma_{SL}x_{SL}} \tag{40}$$

With 15, 26 and 13 is possible to write the movement equation.

$$Dgradc \cdot \mathbf{m} = (1 - c_{SL})\dot{m} \tag{41}$$

Replacing $\dot{m}$ by 40.

$$Dgradc \cdot \mathbf{m} = (1 - c_{SL}) \frac{RT}{\beta M_{SL}} ln\frac{\gamma^e_{SL}x^e_{SL}}{\gamma_{SL}x_{SL}} \tag{42}$$

When the concentration of solid ions in liquid is next to equilibrium concentration, is possible to consider the approach proposed by Lehner and Bataille (1984).

$$Dgradc \cdot \mathbf{m} = K(1 - c_{SL})(1 - c_{SL}/c^e_{SL}) \tag{43}$$

Where $K = (RT)/(\beta M_{SL})$. When $\frac{K}{D} \gg gradc \cdot \mathbf{m}$ the solute concentration in the interface is approximately equal the equilibrium. Then, on equation 41 we can substitute c by c^e. In this work we assume that $c^e = c_{sat}$, where c_{sat} is the saturation concentration. Despite solubility be a property related to equilibrium, our system is not in equilibrium. Then, the motion equation of the interface is:

$$Dgradc \cdot \mathbf{m} = (1 - c_{sat})V \tag{44}$$

2.5 Results

Hence, solving the equations, 37, 36, 38 and 44, we find the velocity with which the interface moves and it is possible to know how stress is related to solubility.

2.5.1 Saturation Concentration - c_{sat}

To solve the equation 38 on boundary equation is that on the interface solid-solution the concentration is equal the saturation concentration. At articles (Nguyen *et al.*, 2017), (Scheiner and Hellmich, 2007), (Scheiner and Hellmich, 2009) and (Mai and Soghrati, 2017) c_{sat} is a constant value. We propose an equation to c_{sat} when we introduce the configurational force balance and show that c_{sat} depend of stresses on the body.

We use the Thermodynamics of Continua with configurational forces balance to obtain 33. The articles (Lehner and Bataille, 1984) and (Paterson, 1973) propose that stresses influence the solubility, then like proposed by them we introduce the equation 35 to verify that. To verify the influence of stress tension on dissolution, we exclude the hydrostatic effect of fluid pressure. Then we write the chemical potential of a solution in local equilibrium with the solid subject to hydrostatic pressure (μ^p_{SL}). We subtract this value of μ^p_{SL} from

$$\gamma_{SL} x_{SL} = \gamma^p_{SL} x^p_{SL} e^{\left(\frac{(\Delta\psi_S + p\Delta\nu_S) M_{SL}}{RT}\right)}, \tag{45}$$

where,

$$\Delta\psi_S = \psi_S - \psi^p_S \tag{46}$$

$$\Delta\nu_S = 1/\rho_S - 1/\rho^p_S \tag{47}$$

$$\psi_S = \frac{v^0_S}{2E}(\sigma_1^2 + \sigma_2^2 + \sigma_3^2 - 2\nu(\sigma_1\sigma_2 + \sigma_2\sigma_3 + \sigma_1\sigma_3)) \tag{48}$$

$$v_S - v^0_S = v^0_S \left(\frac{1 - 2\nu}{E}\right)(\sigma_1 + \sigma_2 + \sigma_3) \tag{49}$$

where v^0_S is the specific solid volume without stress and ν is the Poisson coefficient.

How in (Lehner and Bataille, 1984) $\frac{\gamma_{SL} x_{SL}}{\gamma^p_{SL} x^p_{SL}} \approx \frac{c_{sat}}{c_p}$, where c_p is the concentration on interface when the solid is subject only to hidrostatic pressure and c_{sat} the concentration when the solid is subject to stresses without hydrostatic pressure. Then,

$$\frac{\gamma_{SL} x_{SL}}{\gamma^p_{SL} x^p_{SL}} = \frac{c_{sat}}{c_p} = e^{\left(\frac{(\Delta\psi_S + p\Delta\nu_S) M_{SL}}{RT}\right)} \tag{50}$$

This equation allow us describe c_{sat}.

2.5.2 Dissolution Rate

The dissolution rate (V) is given by 44.

$$V = \frac{Dgradc \cdot \mathbf{m}}{(1 - c_{sat})} \tag{51}$$

3. NUMERICAL RESULTS

As example we consider a cylinder linear elastic, isotropic, and homogeneous immersed in a stationary solution that causes an external pressure p_e (Figure 3). It is submitted to internal pressure p_i and is undergoing a processes dissolution in its outer diameter. We use COMSOL Multiphysics to solve the diffusion problem in a cylinder, where the equation is:

$$\frac{\partial c}{\partial t} = \frac{1}{r}\frac{\partial}{\partial r}\left(rD\frac{\partial c}{\partial r}\right) \tag{52}$$

$$c(R_e, t) = c_{sat} \tag{53}$$

$$c(\infty, t) = 0 \tag{54}$$

$$c(R_e, 0) = 0 \tag{55}$$

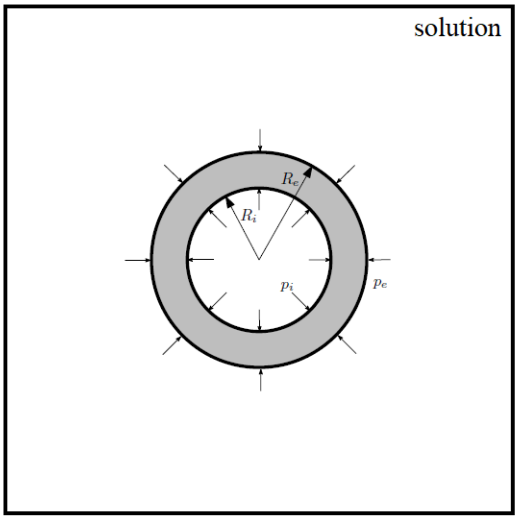


Figure 3. Cylinder immersed in a solution.

Replacing the principal stresses on equation 50 we find the c_{sat}. Plotting this solution, we can compare how the saturation concentration change with internal pressure for different relations between thickness and intern radius. It is possible to observe how much smaller the tube thickness, the greater c_{sat} when the relation between internal and external pressure increases. (Figure 4).

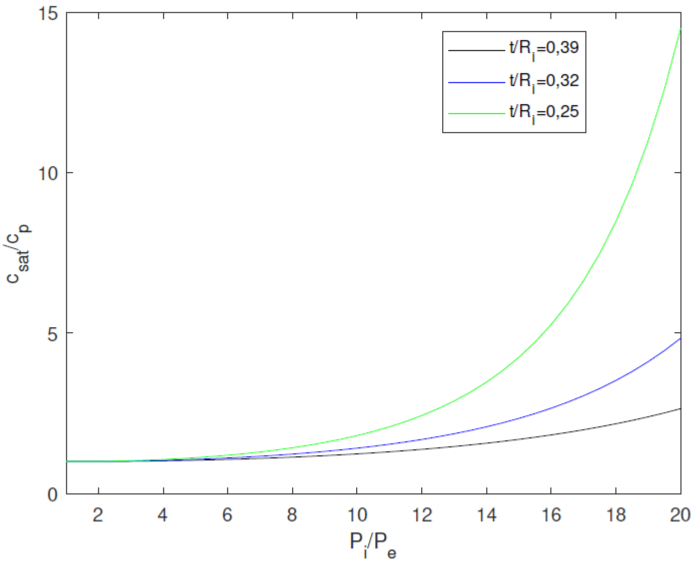
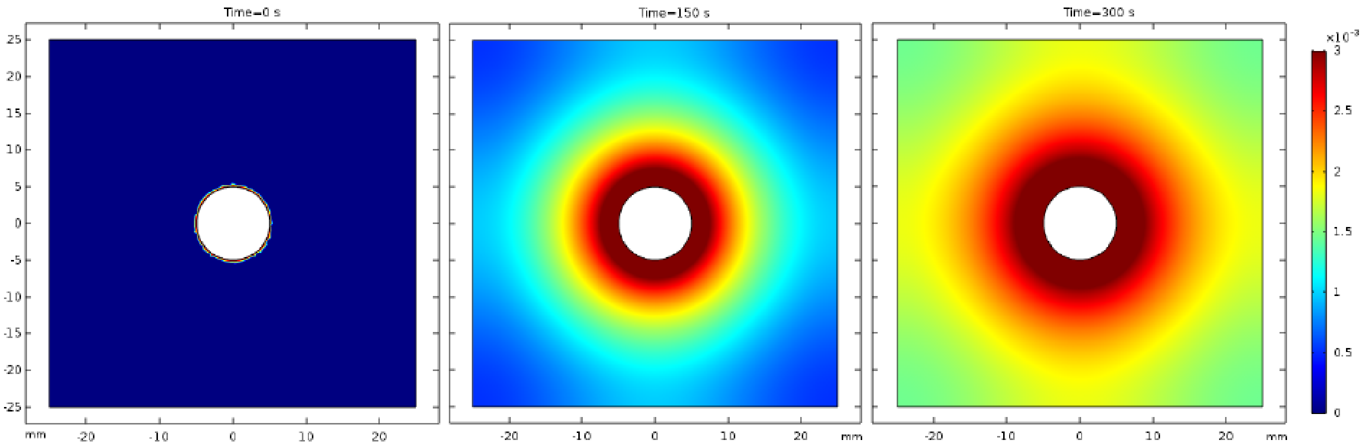


Figure 4. Saturation concentration variation



(a) (b) (c)

Figure 5. (a) t=0s; (b) t= 150s; (c) t=300s

In Figure 5 we can observe the diffusion evolution. With c_{sat} values, it is possible to solve the diffusion problem. And in 5 we have at the initial time the c_{sat} only in the edge, after some time this concentration evolves others parts.

In Figure 6 we can verify how the internal pressure influence at dissolution rate. As the internal pressure increases the dissolution becomes faster. This plot was done using MATLAB. Through this example, it is possible to have an idea of how the stresses influence dissolution.

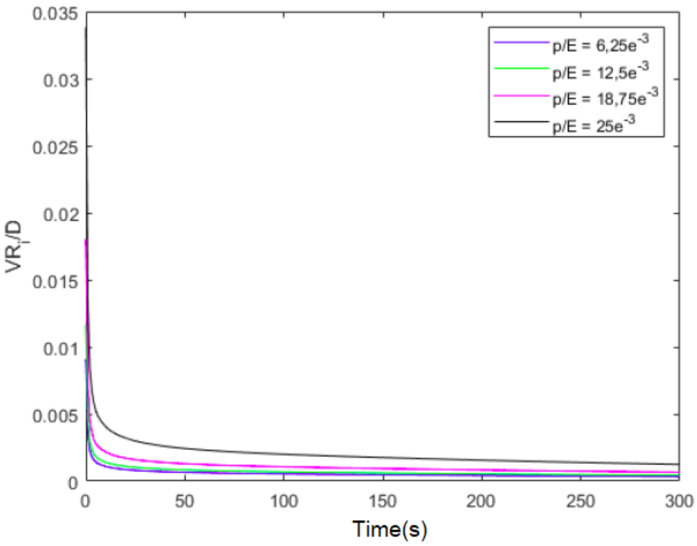


Figure 6. Dissolution rate graph.

4. SUMMARY

The paper presents an equation for calculating the saturation concentration and the introduction of mechanical effects that are capable of influence the dissolution process. Mechanical effects were coupled with solubility through the introduction of the balance of configurational forces and the problem was structured based on Continuum Thermodynamics. Through the use of the balance of configurational forces we have obtained the necessary extra condition at the interface, that is, the saturation concentration, as we could see in the example presented

Next steps, other effects that influence the dissolution process can be included in the presented model. For example, showing equivalence between models of phase field with well-defined interface models. By adjusting the parameters of these models, it will be possible to include the mechanical effects also in the phase field model and then simulate the problem using this computationally more advantageous model.

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